University of Virginia

STRUCTURE AND DYNAMICS OF EXCITED ATOMS

Final Technical Report
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by

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I. Objectives

The main objective of this research is to explore the properties of a cold gas of highly excited, or Rydberg, atoms. This physical system is at the intersection of atomic, plasma, and condensed matter physics and offers the possibility of studying condensed matter and plasmas in unprecedented ways.

The atoms are cold, ~300 μK, so they have typical velocities of 30 cm/s, which means that they move $0.3\mu m$ in the $1\mu s$ duration of a typical experiment. The atoms have a maximum density of 10^{10} cm^{-3}, in which case the typical spacing between atoms is $5\mu m$. Thus, even at the highest densities employed, the atoms move ~5% of the spacing between atoms during a $1\mu s$ experiment, so the atoms are effectively frozen in place, as in an amorphous solid.

In spite of their low density the atoms interact with each other because the transition dipole moments scale as n^2, where n is the principal quantum number. For a gas of $n = 30$ atoms at a density of 10^{10} cm^{-3} the strength of the dipole-dipole interaction between atoms with dipole moments μ_1 and μ_2 separated by R is $\mu_1\mu_2/R^3 = 6$ MHz, which is comparable to the thermal energy of the atoms, i.e. 20 MHz at $T = 300\mu K$. As a result, it is possible to observe phenomena in dilute Rydberg samples which are observed in normal solids at densities of 10^{23} cm^{-3}. For example, we have observed excitation diffusion analogous to spin diffusion in a glass.[1,2] What makes the cold Rydberg gas so interesting is that it is possible to not only observe the interactions but to turn them on and off in real time as well.

In the cold Rydberg gas at 10^{10} cm^{-3} the spacing is $5\mu m$, which can be compared to the orbital radius of the Rydberg atom, which scales as n^2. For $n = 200$ the orbital radius is $5\mu m$, so one would reasonably expect to see something analogous to a Mott insulator-metal transition at this point. However, at far lower n, $n \sim 70$, atoms at 10^{10} cm^{-3} become a plasma in less than

~100*ns* after laser excitation to a Rydberg state.[3] This plasma is extraordinarily cold ~1 – 10*K*, far colder than a textbook cold plasma, which has $T = 10{,}000K$.[4]

The cold Rydberg gas is an ideal model system for the study of interatomic interactions normally observed in solids. What makes this system so fascinating is that interactions can be effectively turned on or off by tuning levels into or out of resonance, which is easily done by the application of quite modest electric fields. In addition the geometry of the sample can be controlled easily. Samples having one, two, or three dimensions are easily created, and the orientation of a sample relative to the atomic dipoles can be controlled as well. Finally, it should be possible to create samples which have either random or regular spacing. To date only random samples have been made.

The fascinating aspect of the plasmas which may be formed from these samples is that the initial conditions are controlled to an extent simply not possible in most plasma physics experiments.[5] Thus quite subtle effects can be explored. For example a mixture of cold ions, electrons with well defined energy and Rydberg atoms with well defined binding energy can be formed. How does it evolve?

Finally cold Rydberg gases have been suggested as a possible way of making fast gates for quantum computing.[6,7] In particular, the proposed approaches employ the dipole-dipole interactions discussed above.

II. Status of Effort

We have now developed the laser technology to excite Rydberg atoms in the trap using a pulsed amplified cw laser. With this system we can produce samples with only 10% fluctuation in the number of Rydberg atoms. This stability is crucial in studying the processes of interest,

which always involve interactions among Rydberg atoms and are thus non linear, often highly so, in the density.

We have developed millimeter (mm) wave techniques to probe and manipulate the cold Rydberg samples. As will be described below, we are able to use the high spectral resolution to observe line broadening, and we have been able to change the macroscopic properties of the sample by changing the quantum state of <10% of the Rydberg atoms.

We have not yet done much to control the geometry of the sample, but it is becoming clear that this is an interesting direction to explore.

III. Accomplishments

Millimeter waves

We have developed millimeter wave spectroscopy as a very sensitive spectral probe with which we can observe very subtle energy shifts and control the properties of the sample.

In our first experiments we found that single photon microwave transitions had linewidths of 5 MHz, due to the inherent B field inhomogeneity of our magneto optical trap. More interesting, we observed transform limited linewidths as low as 30kHz for two photon transitions of the type $n\ell_j \rightarrow (n+1)\ell_j$, in spite of the trap's 10 G/cm magnetic field inhomogeneity.[8] The narrow linewidth comes from the fact that the g_j factors of the two states are the same and all $\Delta m_j = 0$ transitions occur at the same frequency. The $\Delta m_j \neq 0$ transitions occur over a broad range, ~5 MHz, of frequencies and are essentially invisible at low microwave power. We first made spectroscopic measurements of the $\Delta n = 1$ intervals of the *ns*, *np*, and *nd* series, improving the quantum defects by more than an order of magnitude, which will be important if such transitions are used as secondary frequency or wavelength standards, as has been proposed.[9]

We have recently been turning off the inhomogeneous trap field and taking advantage of the fact that the atoms remain trapped in optical molasses.[10] With this technique we are able to observe single photon microwave transitions with linewidths of <1 MHz, which is more than adequate to probes interactions among the atoms.

High Resolution Pulse Amplified cw Laser System

During this period we brought into operation a pulse amplified cw 960nm Ti:Sapphire laser to replace our pulsed dye laser for excitation of the Rydberg states. The shot to shot frequency fluctuations of the dye laser led to enormous, >50%, shot to shot fluctuations in the Rydberg atom population. Such large fluctuations make the study of processes non linear in the number of Rydberg atoms difficult at best. We amplify the 960*nm* light in a two stage dye amplifier and frequency double the pulses to 480*nm*. The result is 480*nm* pulses at a 20Hz repetition rate with pulse duration 10*ns*, pulse energy 20μJ, linewidth 200MHz, and shot to shot intensity fluctuations of <10%. This leads to very reproducible Rydberg atom signals, allowing much more sophisticated experiments.

Many Body Interactions

In our first dipole-dipole energy transfer experiment we observed the resonant energy transfer process[2]

$$Rb\ 25s + Rb\ 33s \rightarrow Rb\ 24p + Rb\ 34p \ , \qquad (1)$$

which was tuned into resonance at the fields of 3.0 and 3.4 V/cm. Much of the linewidth of the observed resonances was attributed to the processes

$$Rb\ 25s + Rb\ 24p \rightarrow Rb\ 24p + Rb\ 25s \qquad (2)$$

and

$$Rb\ 33s + Rb\ 34p \rightarrow Rb\ 34p + Rb\ 33s \qquad (3)$$

which are always resonant. Since the processes of Eqs. (2) and (3) were always present, it was not possible to verify how they affected the linewidth of the process of Eq. (1) in a clear way.

We have verified the importance of the always resonant processes of Eqs. (2) and (3) by two different experiments. The first experiment was a Ramsey interference experiment in which we brought the atoms into resonance at $E = 34$ V/cm with two $100ns$ long field pulses separated by a variable time T.[11] During time T the field is off resonant and thus the interaction of Eq. (1) is effectively turned off, but those of Eqs. (2) and (3) are still present. We observed interference fringes as T was scanned, and the fringes exhibited a density dependent decay rate due to the inhomogeneity of the interaction strengths of Eqs. (2) and (3) across the sample.

The second approach we took was to explicitly introduce an interaction analogous to those of Eqs. (2) and (3) but independent of the energy transfer of Eq. (1).[12] Explicitly, we introduced atoms in the $34s$ state with a microwave pulse which drives the two photon $33s \rightarrow 34s$ transition and introduces the process

$$Rb\ 34s + Rb\ 34p \rightarrow Rb\ 34p + Rb\ 34s \qquad (4)$$

which is independent of the process of Eq. (1). We have shown that the process of Eq. (4) does indeed broaden the resonances of Eq. (1). What is somewhat surprising is that it also increases the peak resonance signal. Normally one would expect the area of the resonances to remain constant. The combination of broader and stronger resonance signals is a clear indication of the fact that the interaction among all the participating atoms is coherent.

Plasma Formation

One of the more surprising discoveries in this program was that a cold dense sample of Rydberg atoms spontaneously evolves into a plasma.[3] The evolution occurs in the following way if we start with $n = 35$ atoms.. Ionization of the atoms occurs because of black body

radiation induced photoionization and collisions between cold Rydberg atoms and the hot 300K background atoms. After a few percent of the cold Rydberg are ionized, their macroscopic positive charge traps subsequently liberated electrons which pass back and forth through the cloud of Rydberg atoms leading to collisional avalanche ionization.

In the first experiment there were two puzzling results.[3] First, where does the energy come from to ionize the atoms? If the atoms are all in the $n = 40$ state at the beginning each atom which is ionized gains at least 70 cm^{-1} of energy. What is its origin? Second, for $n > 50$ initial states the evolution to a plasma was very rapid even if there were no hot 300K atoms. Since the black body indication photoionization rates decrease with n this observation was puzzling.

The energy question was resolved by making careful measurements of the total number of ions plus Rydberg atoms *vs* time after laser excitation.[13] We found that the number decreased with time (~30% in 10μs) and that the rate of decrease increased with initial number density. The loss in the number of atoms and ions is due to atoms' making transitions to low lying states not ionized by the field ionization pulse. This notion was reinforced by adjusting the amplitude of the field ionization pulse to alter the value of n above which we ionized the atoms. With larger field ionization pulses the observed number of atoms and ions did not decrease as rapidly. These observations imply that during the collisional avalanche some electrons drive atoms to lower states so that the electrons gain energy from these collisions. This electron energy is subsequently given to other atoms, which are ionized by electron collisions.

The second mystery was the rapid plasma formation at high n. We have recently discovered that it is due to the almost resonant dipole-dipole interaction. Specifically, for $n > 40$ there exist nearly resonant dipole-dipole couplings even with no applied electric field.

Consequently half of the most closely spaced pairs of atoms are attracted to each other and collide to produce the initial ionization needed to seed the avalanche.[14] A particularly graphic demonstration of this phenomena was based on the microwave transition from a pair of excited *s* atoms, which have minimal interaction, to an *sp* pair which has strong attractive and repulsive dipole-dipole interactions. On the attractive side of the *s-p* microwave transition ionization occurred, but on the repulsive side nothing happened, verifying that the dipole-dipole interaction does produce the initial ionization.

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14. W. Li, P. Tanner, and T.F. Gallagher (unpublished).

V. Personnel Supported

During this period Thomas F. Gallagher (PI), Igor Mourachko (post doctoral fellow), Wenhui Li (graduate student), and Paul Tanner (graduate student) were supported by the AFOSR grant. Michael Robinson, supported by the Palace Knight program, played an important role in the work.

VI. Publications

1. W.R. Anderson, M.P. Robinson, J.D.D. Martin, and T.F. Gallagher, "Dephasing of resonant energy transfer in a cold Rydberg gas," Phys. Rev. A **65**, 063404 (2002).

2. T.F. Gallagher, M.P. Robinson, B. Laburthe-Tolra, M.W. Noel, and P. Pillet, "Evolution of Cold Rydberg Atoms into an Ultracold Plasma," in *Atomic Processes in Plasmas*, ed. By D.R. Schultz, F.W. Meyer, and F. Ownby (American Institute of Physics, New York, 2002).

3. W. Li, I. Mourachko, M.W. Noel, and T.F. Gallagher, "Millimeter wave spectroscopy of cold Rb atoms in a magneto-optical trap: Quantum defects of the ns, np, and nd Series," Phys. Rev. A **67**, 052502 (2003).

4. T.F. Gallagher, P. Pillet, M.P. Robinson, B. Laborthe-Tolra, and M.W. Noel, "Back and Forth between Rydberg Atoms and ultracold plasmas," J. Opt. Soc. Am. B **20,** 1091 (2003).

5. I. Mourachko, W. Li, and T.F. Gallagher, "Controlled many-body interactions in a frozen Rydberg gas," Phys. Rev. A **70,** 031401 (2004).

6. W. Li, M.W. Noel, M.P. Robinson, P.J. Tanner, T.F. Gallagher, D. Comparat, B. Laburthe-Tolra, N. Vanhaecke, T. Vogt, N. Zahzam, P. Pillet, and D.A. Tate, "Evolution dynamics of a dense frozen Rydberg gas to plasma," Phys. Rev. A **70,** 042713 (2004).

VII. Interactions

a. Participation:

Invited paper, "Evolution of cold Rydberg Atoms into an Ultracold Plasma," Thirteenth APS Topical Conference on Atomic Processes in Plasmas, Gatlinburg, April 2002.

Invited paper, "From Rydberg Atoms to an Ultra cold Plasma and Back," Cooling 2002, Visby (Sweden), June 2002.

Invited paper, "Rydberg Atoms: A laboratory for Atoms, Plasmas, and Solids," Atomes Exotiques – Chaos, Orsay (France), December 2002.

Seminar, "Evolution from a Cold Rydberg Gas to an Ultracold Plasma," University of Stockholm, Stockholm, (Sweden), June 2002.

Seminar, "Evolution from a Cold Rydberg Gas to an Ultracold Plasma," Max Born Institute, Berlin, September 2002.

Lecturer, International Workshop and Seminar on Rydberg Physics, Dresden, April-May (2004).

Invited paper, "Resonant Dipole Dipole Energy Transfer," Decoherence, entanglement and information protection in complex quantum systems, Les Houches (France) April, 2004.

Seminar, "Dipole-Dipole Interactions in a Frozen Rydberg Gas," Peking University, Beijing, July, 2004.

Invited paper, "Dipole-dipole Interactions in a Frozen Rydberg Gas," First International Symposium on Cold Atom Physics" Lushan (China) July, 2004

b. Consulting: none

c. Transitions: none

VIII. Inventions: none

IX. Awards

Fellow of the American Physics Society

Fellow of the Optical Society of America

Davisson-Germer Prize of the American Physical Society 1996

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